

Evaluation of PM_{2.5} surface concentration simulated by Version 1 of the NASA's MERRA Aerosol Reanalysis over Israel and Taiwan

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Abstract

Version 1 of the NASA MERRA Aerosol Reanalysis (MERRAero) assimilates bias-corrected aerosol optical depth (AOD) data from MODIS-Terra and MODIS-Aqua, and simulates particulate matter (PM) concentration data to reproduce a consistent database of AOD and PM concentration around the world from 2002 to the end of 2015. The purpose of this paper is to evaluate MERRAero's simulation of fine PM concentration against surface measurements in two regions of the world with relatively high levels of PM concentration but with profoundly different PM composition, those of Israel and Taiwan. Being surrounded by major deserts, Israel's PM load is characterized by a significant contribution of mineral dust, and secondary contributions of sea salt particles, given its proximity to the Mediterranean Sea, and sulfate particles originating from Israel's own urban activities and transported from Europe. Taiwan's PM load is composed primarily of anthropogenic particles (sulfate, nitrate and carbonaceous particles) locally produced or transported from China, with an additional contribution of springtime transport of mineral dust originating from Chinese and Mongolian deserts. The evaluation in Israel produced favorable results with MERRAero slightly overestimating measurements by 6% on average and reproducing an excellent year-to-year and seasonal fluctuation. The evaluation in Taiwan was less favorable with MERRAero underestimating measurements by 42% on average. Two likely reasons explain this discrepancy: emissions of anthropogenic PM and their precursors are largely uncertain in China, and MERRAero doesn't include nitrate particles in its simulation, a pollutant of predominately anthropogenic sources. MERRAero nevertheless simulates well the concentration of fine PM during the summer, when Taiwan is least affected by the advection of pollution from China.

Keywords: MERRAero, Evaluation, Fine particulate matter, Israel, Taiwan.

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INTRODUCTION

NASA's Modern-Era Retrospective Analysis for Research and Application (MERRA, Rienecker *et al.*, 2011) is a reanalysis tool integrating satellite observations from the Earth Observing System and model data from the 5th version of the Goddard Earth Observing System (GEOS-5) atmospheric model and data assimilation system (Rienecker *et al.*, 2008) in order to produce a consistent database in both time and space of various environmental variables around the world since the beginning of the satellite era. Recently, bias-corrected aerosol optical depth (AOD) observations from the Moderate Resolution Imaging Spectroradiometers (MODIS, Remer *et al.*, 2005) on board the Terra and Aqua satellites as well as the Goddard Chemistry, Aerosol, Radiation and Transport (GOCART) model (Chin *et al.*, 2002) were included in MERRA to create a reanalysis of aerosols labelled "MERRAero". GOCART simulates the sources, sinks, transport and concentration of sulfate (SO₄), organic carbon (OC), black carbon (BC), dust (DS) and sea salt (SS) aerosols (Chin *et al.*, 2002; Colarco *et al.*, 2010). DS and SS emissions are a function of surface properties and wind speed at the surface, and their respective concentrations are classified in different diameter bins. Sources of other species are simulated from emission inventories, including their precursors. Sulfur dioxide (SO₂, the precursor of SO₄) anthropogenic emissions are input from the Emission Database for Global Atmospheric Research (EDGAR) version 4.1 inventory from 2005 and biomass burning emissions (primarily OC and BC) are input from the NASA Quick Fire Emission Dataset (QFED) version 2.1 (Buchard *et al.*, 2015).

MERRAero simulates the concentration of the five aerosol species listed in the previous paragraph all over the world with a resolution of 0.5° latitude by 0.625° longitude and 72 vertical layers (from the surface to 80 km) from 2002 to the end of 2015 (Buchard *et al.*, 2015). Considering that these aerosol species, also referred to airborne particulate matter (PM), affect public health and visibility differently

63 depending on their size and chemistry (e.g., Laden *et al.*, 2000; Schwartz and Neas, 2000; Groblicki *et*
64 *al.*, 1981), MERRAero's differentiation of the aerosols' chemical speciation is a significant improvement
65 for studying a broad range of air quality issues around the world since very few monitoring networks
66 make such a distinction of local PM observations, but especially in regions with unreliable or scarce
67 monitoring.

68 Different components of MERRAero have been evaluated in different regions of the world. Its
69 assimilation of AOD has been validated over Africa, South America, central and eastern Asia using many
70 remote sensing instruments (Buchard *et al.*, 2015); in the United States, the surface concentrations of
71 PM_{2.5}, their chemical speciation and SO₂ has been thoroughly evaluated (Buchard *et al.*, 2014; 2016);
72 and in Europe, an evaluation of the surface concentrations of PM₁₀, PM_{2.5} and some of their chemical
73 speciation has been performed (Provençal *et al.*, 2016). The concentrations of PM₁₀, PM_{2.5} and SO₄ were
74 generally well simulated in both the U.S. and Europe but Buchard *et al.* (2016) and Provençal *et al.*
75 (2016) noticed an underestimation of carbonaceous concentration in urban/suburban locations,
76 particularly in winter, due to unresolved sources by MERRAero.

77 The U.S. and Europe have similar PM signatures in the sense that both regions are highly
78 industrialized and therefore anthropogenic particles contribute significantly to their PM load. At the same
79 time, implementation of air quality regulation has successfully reduced the emissions of various
80 atmospheric pollutants across the U.S. and Europe over the last decades (e.g. Granier *et al.*, 2011;
81 Klimont *et al.*, 2013; Hand *et al.*, 2012; Xing *et al.*, 2013; de Gouw *et al.*, 2014; Vestreng *et al.*, 2007)
82 and, as a result, maintained relatively low levels of PM concentration. There are nevertheless important
83 differences with respect to the chemical speciation of PM between the two regions such as a
84 predominance of carbonaceous particles over the western U.S. due to summer wildfires and a
85 predominance of dust particles over southern Europe due to its proximity to the Sahara desert.

86 The PM signature in the U.S. and Europe is not representative of many other regions in the world
87 where PM sources and pollution control are profoundly different. In order for MERRAero to achieve
88 optimal reliability for studying air quality issues around the world, the purpose of this article is to pursue
89 MERRAero's evaluation in regions with different and distinct aerosol signatures, those of Israel and
90 Taiwan. The evaluation in Israel, a region with a heavy PM load due to its proximity to major deserts,
91 will ascertain MERRAero's ability to simulate the concentration of aerosol originating from natural
92 sources. Taiwan being located in a region of the world which is routinely experiencing severe air
93 pollution episodes, the evaluation there will provide insight on MERRAero's applicability in highly
94 polluted regions where its contribution would be most beneficial.

95

96 **LOCATIONS AND METHODS**

97

98 *Israel and Taiwan*

99 Israel is located in western Asia, surrounded by the Mediterranean Sea, the Sahara desert and the
100 Middle Eastern deserts. Its PM concentration load is relatively high, composed largely of mineral DS
101 (Kushelevsky *et al.*, 1983; Malenky *et al.*, 1983; Foner and Ganor, 1992) with an occasionally important
102 contribution from SS particles when the wind is blowing inland (Foner and Ganor, 1992). PM
103 concentration in urban areas such as the coastal city of Tel Aviv is even higher due to anthropogenic SO₄
104 locally produced or transported from Europe (Foner and Ganor, 1992). Rural locations in Israel have also
105 been impacted by the advection of SO₄ particles originating from Europe (Luria *et al.*, 1989).

106 Taiwan is an island located in eastern Asia, separated from mainland China by the Taiwan Strait.
107 Its concentration level of PM is fairly high, especially in urban areas (Chen *et al.*, 1999), caused by
108 industrial and transportation activities within Taiwan but also due to wintertime synoptic features that

109 transport polluted air from China (Lin *et al.*, 2005). SO₄, OC, BC, nitrate (NO₃) and ammonium (NH₄)
110 particles together compose a large portion of PM concentration (Lin *et al.*, 2008; Lin, 2002; Chen *et al.*,
111 2003; Tsai and Kuo, 2005; Tsai and Cheng, 1999; 2004). Taiwan nevertheless enjoys cleaner air during
112 the summer, coinciding with the typhoon season which sweeps the island with strong winds and heavy
113 rain (Lin *et al.*, 2008). In spring, Taiwan is also impacted by the advection of dust originating from
114 Chinese and Mongolian deserts (Chen *et al.*, 2004).

115

116 ***Evaluation method***

117 MERRAero simulates the concentration of five PM_{2.5} (PM with diameter ≤ 2.5 μm) species every
118 hour: SO₄, OC, BC, DS_{2.5} and SS_{2.5}. From these, it is possible to apply a mass reconstruction method to
119 estimate the total concentration of PM_{2.5}. Chow *et al.* (2015) reviewed 11 commonly used equations to
120 reconstruct PM mass from speciation measurements which are usually determined by the measurements
121 available. The equations usually took the following form:

122

$$123 \quad \text{PM} = \text{Inorganic ions} + \text{Organic matter} + \text{BC} + \text{DS} + \text{SS} \quad (1)$$

124

125 Inorganic ions include SO₄, NO₃ and NH₄ ions. When NH₄ measurements were lacking, SO₄ and
126 NO₃ were assumed to be fully neutralized by NH₄ in the form of ammonium sulfate ((NH₄)₂SO₄) and
127 ammonium nitrate (NH₄NO₃) by multiplying their respective concentrations by 1.375 and 1.29;
128 (NH₄)₂SO₄ being composed of 73% of SO₄ by mass and NH₄NO₃ being similarly composed of 78% of
129 NO₃. The concentration of inorganic ions was ultimately estimated by: $1.375 \times [\text{SO}_4] + 1.29 \times [\text{NO}_3]$
130 (brackets denote concentration). [NH₄NO₃] was occasionally omitted altogether when NO₃
131 measurements were lacking or unreliable (e.g., Malm *et al.*, 1994).

132 The concentration of particulate organic matter (POM) was estimated through OC measurements
 133 multiplied by a coefficient which took into account other organic compounds found in POM but not
 134 measured. Commonly and historically, a coefficient of 1.4 was used (Chow *et al.*, 2015; Turpin and Lim,
 135 2001), but Turpin and Lim (2001) argued that such a value is often too low. They recommended a value
 136 of 1.6 ± 0.2 for urban carbonaceous particles, 2.1 ± 0.2 for aged (non-urban) particles and a value as high
 137 as 2.6 for biomass burning particles.

138 Taking into consideration the PM species simulated by MERRAero and given that this evaluation
 139 is performed in a combination of urban and non-urban locations, the following reconstruction is used:

140

$$141 \quad [PM_{2.5}] = 1.375 \times [SO_4] + 1.8 \times [OC] + [BC] + [DS_{2.5}] + [SS_{2.5}] \quad (2)$$

142

143 Eq. 2 lacks the concentration of NO_3 particles whose sources are predominantly anthropogenic in nature
 144 (Delmas *et al.*, 1997).

145 MERRAero's simulation at the surface is compared to hourly observations of $[PM_{2.5}]$ measured at
 146 11 locations in Israel between 2003 and 2014, and 13 locations in and around Taiwan between 2005 and
 147 2014 (Fig. 1). A spatial consistency algorithm is applied to assure reliability of the observed and
 148 simulated data which goes as follows: since trace concentrations are usually lognormally distributed, the
 149 bias between log-simulated concentration and log-observed concentration ($B_{log} = \log(C_s) - \log(C_o)$; C_s :
 150 simulated concentration, C_o : observed concentration) is calculated at all locations within each study areas
 151 on a given hour; the average and standard deviation of B_{log} are calculated and used to define a reliability
 152 interval which justifies ~95% of the normal distribution: $\overline{B_{log}} \pm 2\sigma_{B_{log}}$; all data pairs that fall outside
 153 this interval are excluded.

154 Performance statistics are calculated to quantify MERRAero's accuracy: the mean fraction
 155 $\overline{F} = \overline{C_s} / \overline{C_o}$, the mean bias $\overline{B} = \overline{C_s} - \overline{C_o}$, the standard deviation of the bias (SD-B) and the
 156 correlation coefficient (R). Given that trace concentrations are lognormally distributed, it is also relevant
 157 to compute log-transformed statistics: $\overline{B_{log}}$, SD- B_{log} and R_{log} . Willmott (1982) criticized the use of R to
 158 evaluate model performance since it doesn't directly compare simulated with observed data. Therefore,
 159 Chang and Hanna (2004) recommended as a rigorous index to evaluate air quality models the proportion
 160 of simulated data which falls within a factor of 2 of observed data (FAC2, i.e. proportion of the data
 161 which satisfies $0.5 \leq C_o / C_s \leq 2.0$) since this index is not disproportionately sensitive to extreme values
 162 and is unaffected by simplification of errors. Chang and Hanna (2004) considered a model's performance
 163 to be reasonably good if $FAC2 \geq 0.5$.

164

165 **RESULTS AND DISCUSSION**

166

167 ***Israel***

168 The spatial consistency algorithm excluded 5% of the data in Israel. At $22.5 \mu\text{g m}^{-3}$, the $PM_{2.5}$ load
 169 in Israel is high (Table 1) compared to Europe (Provençal *et al.*, 2016), rural and suburban U.S. (Buchard
 170 *et al.*, 2016). Overall, MERRAero simulates $[PM_{2.5}]$ very well in Israel by slightly overestimating its
 171 average concentration by 6% or $1.4 \mu\text{g m}^{-3}$ (Table 1). However, the high SD-B value and modest R
 172 suggest significant scatter within the data and a low bias resulting from simplification of errors. On the
 173 other hand, it is worth mentioning that SD's are disproportionately impacted by extreme data pairs. For
 174 instance, if such data which fall outside a factor of 5 between observed and simulated concentrations,
 175 which represent 2.6% of the sample, \overline{B} and SD-B are reduced to $1.0 \mu\text{g m}^{-3}$ and $19.6 \mu\text{g m}^{-3}$,

176 respectively. Furthermore, the density scatter plot of Fig. 2a reveals that although there is some scatter,
177 the bulk of the data is generally well simulated. This is further supported by a high FAC2 value of 76%.
178 The log-transformed data (Table 1; Fig. 2b) support a similar analysis. Additionally, Fig. 3 compares the
179 annual and monthly fluctuations between simulated and observed data, and illustrates an excellent
180 identity between both datasets.

181 MERRAero's ability to accurately estimate $[PM_{2.5}]$ in Israel relies predominantly on its ability to
182 simulate $[DS_{2.5}]$ since $[PM_{2.5}]$ is largely composed of this species (Table 2) and, to a lesser extent, its
183 ability to simulate $[SO_4]$ and $[SS_{2.5}]$. The evaluation of $[DS_{2.5}]$ in the U.S. revealed important seasonal
184 biases without much impact on the evaluation of $[PM_{2.5}]$ given its small contribution to $[PM_{2.5}]$ there
185 (Buchard *et al.*, 2016). While the U.S. is mostly impacted by long range transport of DS, this evaluation
186 in Israel would suggest that MERRAero performs well in simulating $[DS_{2.5}]$ originating from local
187 sources. $[SO_4]$ has been shown to be well simulated in the U.S. and in Europe (Buchard *et al.*, 2016;
188 Provençal *et al.*, 2016), we therefore have no reason to believe otherwise in this region. MERRAero
189 largely overestimated $[SS_{2.5}]$ in both the U.S. and Europe due in part to measurement biases which could
190 very well be the case in Israel. An overestimation of $[SS_{2.5}]$ could compensate the lack of nitrate particles
191 in the simulation. In any case, the lack of nitrate particles is likely a minor shortcoming given that they
192 are less abundant than SO_4 and probably contribute little to $[PM_{2.5}]$.

193

194 ***Taiwan***

195 The spatial consistency algorithm excluded 3% of the data in Taiwan. The $PM_{2.5}$ load in Taiwan is
196 higher than in Israel ($29.8 \mu g m^{-3}$; Table 1). Despite a FAC2 of 59%, MERRAero's performance in
197 Taiwan is much less encouraging. On average, MERRAero underestimates total $[PM_{2.5}]$ by $8.8 \mu g m^{-3}$,
198 a factor of 1.42. The SD-B is also high and R is positive but low. Fig. 4a–b reveals that the bulk of the

199 simulated data, 66% to be precise, is indeed underestimated.

200 MERRAero's simulation of [PM_{2.5}] in Taiwan is mostly anthropogenic in nature (Table 2) with a
201 significant proportion attributed to SO₄. This information coupled with Fig. 5 which compares annually
202 and monthly averaged simulated and observed concentration reveals a few clues as to why MERRAero's
203 performance is less favorable in Taiwan. The evaluation performs well during the summer (typhoon)
204 season but deteriorates during the rest of the year when Taiwan is most impacted by the advection of
205 pollution from China. The use of a constant inventory of SO₂ emissions from 2005 is problematic in the
206 long term since it is increasingly becoming antiquated with every passing year. Indeed, in 2005, China
207 successfully implemented comprehensive policies to reduce SO₂ emissions. As a result, SO₂ emissions
208 and, by extension, SO₄ concentrations have been decreasing since 2006 (Lu *et al.*, 2010; 2011; Wang
209 and Hao, 2012; Zhang *et al.*, 2012; Klimont *et al.*, 2013; B. Zhao *et al.*, 2013; Y. Zhao *et al.*, 2013). This
210 is reflected in Fig. 5 with a near constant decrease of [PM_{2.5}] observations as opposed to the near constant
211 year to year concentrations simulated by MERRAero. SO₂ emission estimates from China are also
212 crippled with uncertainties (Smith *et al.*, 2011). The lack of nitrate particles in the simulation is much
213 more troublesome in Taiwan given that [PM_{2.5}] is mostly composed of anthropogenic particles. This
214 would certainly explain a significant portion of the underestimation. Another possible explanation for
215 the wintertime discrepancy, one that's also been highlighted by Provençal *et al.* (2016) for the evaluation
216 in Europe, is local sources of pollution unresolved by MERRAero. While MERRAero's simulation takes
217 into account urban sources of pollution, its resolution is too coarse to capture the urban core of cities.
218 Some monitoring stations in Taiwan (Fig. 1) are located in or around large cities, but none of them are
219 located in their downtown core. We therefore don't expect them to be overly influenced by local sources
220 of pollution. Nevertheless, some influence of unresolved sources should be anticipated. The springtime

maximum observed in Fig. 5 is the likely contribution of long range transport of DS, well captured by MERRAero.

CONCLUSION

We evaluated version 1 of the MERRA Aerosol Reanalysis' ability to simulate the concentration of $PM_{2.5}$ in two regions of the world with relatively high levels of PM concentration but with profoundly different PM composition. Israel is characterized by a high concentration of $PM_{2.5}$ due to its proximity to major deserts and to the highly saline Mediterranean Sea. Its $PM_{2.5}$ load is composed mostly of natural particles (mineral dust and sea salt) with some contribution of anthropogenic particles (sulfate) originating from Israel's urban activities and advection from Europe. Taiwan's high $PM_{2.5}$ concentration is mostly anthropogenic in nature due to Taiwan's own industrial activities and to advection of polluted air from China with some contribution of dust particles originating from east Asian deserts.

The evaluation reproduced favorable results in Israel where MERRAero slightly overestimated actual $PM_{2.5}$ concentration by 6% on average. Although there is scatter within the distribution, most of the simulation is reasonably accurate with over 75% of the simulated data falling within a factor of 2 of measurements. Given that most of $PM_{2.5}$ in Israel is mineral dust, this evaluation supports the assumption that MERRAero performs well in simulating the concentration of fine dust originating from local and regional sources throughout the year.

The evaluation is not as favorable in Taiwan where MERRAero significantly underestimated measured $PM_{2.5}$ concentration by 42% on average. Given that $PM_{2.5}$ in Taiwan is mostly composed of anthropogenic particles, many of which originate from China, two likely reasons explain this outcome: the uncertainty with respect to Chinese emissions and the lack of nitrate particles in the simulation. The

simulation was indeed better during the summer when Taiwan is least impacted by advection of polluted air from China.

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REFERENCES

- Buchard, V., da Silva, A.M., Colarco, P., Krotkov, N., Dickerson, R.R., Stehr, J.W., Mount, G., Spinei, E., Arkinson, H.L. and He, H. (2014). Evaluation of GEOS-5 sulfur dioxide simulations during the Frostburg, MD 2010 field campaign. *Atmos. Chem. Phys.* 14: 1929–1941.
- Buchard, V., da Silva, A.M., Colarco, P.R., Darmenov, A., Randles, C.A., Govindaraju, R., Torres, O., Campbell, J. and Spurr, R. (2015). Using the OMI aerosol index and absorption aerosol optical depth to evaluate the NASA MERRA Aerosol Reanalysis. *Atmos. Chem. Phys.* 15: 5743–5760.
- Buchard, V., da Silva, A.M., Randles, C.A., Colarco, P., Ferrare, R., Hair, J., Hostetler, C., Tackett, J. and Winker, D. (2016). Evaluation of the surface PM_{2.5} in Version 1 of the NASA MERRA Aerosol Reanalysis over the United States. *Atmos. Environ.* 125: 100–111.
- Chang, J.C. and Hanna, S.R. (2004). Air quality model performance evaluation. *Meteorol. Atmos. Phys.* 87: 167–196.
- Chen, K.S., Lin, C.F. and Chou, Y.M. (2003). Determination of source contribution to ambient PM_{2.5} in Kaohsiung, Taiwan, using a receptor model. *J. Air Waste Manage. Assoc.* 51: 489–498.
- Chen, M.L., Mao, T.F. and Lin, I.K. (1999). The PM_{2.5} and PM₁₀ particles in urban areas of Taiwan. *Sci.*

268 *Total Environ.* 226: 227–235.

269 Chen, Y.S., Sheen, P.C., Chen, E.R., Liu, Y.K., Wu, T.N. and Yang, C.Y. (2004). Effects of Asian dust
 270 storm events on daily mortality in Taipei, Taiwan. *Environ. Res.* 95: 151–155.

271 Chin, M., Ginoux, P., Kinne, S., Torres, O., Holben, B.N., Duncan, B.N., Martin, R.V., Logan, J.A.,
 272 Higurashi, A. and Nakajima, T. (2002). Tropospheric aerosol optical thickness from the GOCART
 273 model and comparisons with satellite and sun photometer measurements. *J. Atmos. Sci.* 59: 461–
 274 483.

275 Chow, J.C., Lowenthal, D.H., Antony Chen, L.W., Wang, X. and Watson, J.G. (2015). Mass
 276 reconstruction methods for PM_{2.5}: a review. *Air Qual. Atmos. Health* 8: 243–263.

277 de Gouw, J.A., Parrish, D.D., Frost, G.J. and Trainer, M. (2014). Reduced emissions of CO₂, NO_x, and
 278 SO₂ from U.S. power plants owing to switch from coal to natural gas with combined cycle
 279 technology. *Earth's Future* 2: 75–82.

280 Delmas, R., Serça, D. and Jambert, C. (1997). Global inventory of NO_x sources. *Nutr. Cycl. Agroecosyst.*
 281 48: 51–60.

282 Foner, H.A. and Ganor, E. (1992). The chemical and mineralogical composition of some urban
 283 atmospheric aerosols in Israel. *Atmos. Environ.* 26B: 125–133.

284 Granier, C., Bessagnet, B., Bond, T., d'Angiola, A., van der Gon, H.D., Frost, G.J., Heil, A., Kaiser, J.W.,
 285 Kinne, S., Klimont, K., Kloster, S., Lamarque, J.F., Lioussé, C., Masui, T., Meleux, F., Mieville,
 286 A., Ohara, T., Raut, J.C., Riahi, K., Schultz, M.G., Smith, S.J., Thompson, A., van Aardenne, J.,
 287 van der Werf, G.R. and van Vuuren, D.P. (2011). Evolution of anthropogenic and biomass burning
 288 emissions of air pollutants at global and regional scales during the 1980–2010 period. *Clim. Chang.*
 289 109: 163–190.

290 Groblicki, P.J., Wolff, G.T. and Countess, R.J. (1981). Visibility-reducing species in the Denver “brown

cloud”—I. Relationships between extinction and chemical composition. *Atmos. Environ.* 15: 2473–2484.

Hand, J.L., Schichtel, B.A., Malm, W.C. and Pitchford, M.L. (2012). Particulate sulfate ion concentration and SO₂ emission trends in the United States from the early 1990s through 2010. *Atmos. Chem. Phys.* 12: 10353–10365.

Klimont, Z., Smith, S.J. and Cofala, J. (2013). The last decade of global anthropogenic sulfur dioxide: 2000–2011 emissions. *Environ. Res. Lett.* 8: 014003.

Kushelevsky, A., Shani, G. and Haccoun, A. (1983). Effect of meteorological conditions on total suspended particulate (TSP) levels and elemental concentration of aerosols in a semi-arid zone (Beer-Sheva, Israel). *Tellus* 35B: 55–64.

Laden, F., Neas, L.M., Dockery, D.W. and Schwartz, J. (2000). Association of fine particulate matter from different sources with daily mortality in six U.S. cities. *Environ. Health Persp.* 108: 941–947.

Lin, C.H., Wu, Y.L., Lai, C.H., Watson, J.G. and Chow, J.C. (2008). Air quality measurements from the southern particulate matter supersite in Taiwan. *Aerosol Air Qual. Res.* 8: 233–264.

Lin, C.Y., Liu, S.C., Chou, C.C.K., Huang, S.J., Liu, C.M., Kuo, C.H. and Young, C.Y. (2005). Long-range transport of aerosols and their impact on the air quality of Taiwan. *Atmos. Environ.* 39: 6066–6076.

Lin, J.J. (2002). Characterization of the major chemical species in PM_{2.5} in the Kaohsiung City, Taiwan. *Atmos. Environ.* 36: 1911–1920.

Lu, Z., Streets, D.G., Zhang, Q., Wang, S., Carmichael, G.R., Cheng, Y.F., Wei, C., Chin, M., Diehl, T. and Tau, Q. (2010). Sulfur dioxide emissions in China and sulfur trends in East Asia since 2000. *Atmos. Chem. Phys.* 10: 6311–6331.

Lu, Z., Zhang, Q. and Streets, D.G. (2011). Sulfur dioxide and primary carbonaceous aerosol emissions

314 in China and India, 1996–2010. *Atmos. Chem. Phys.* 11: 9839–9864.

315 Luria, M., Lifschitz, B. and Peleg, M. (1989). Particulate sulfate levels at a rural site in Israel. *J. Atmos.*
316 *Chem.* 8: 241–250.

317 Malenky, B., van Grieken, R., Van’t Dack, L. and Luria, M. (1983). Atmospheric trace element
318 concentration in Jerusalem, Israel. *Atmos. Environ.* 17: 819–822.

319 Malm, W.C., Sisler, J.F., Huffman, D., Eldred, R.A. and Cahill, T.A. (1994). Spatial and seasonal trends
320 in particle concentration and optical extinction in the United States. *J. Geophys. Res.* 99: 1347–
321 1370.

322 Provençal, S., Buchard, V., da Silva, A.M., Leduc, R. and Barrette, N. (2016). Evaluation of PM surface
323 concentrations simulated by Version 1 of the NASA MERRA Aerosol Reanalysis over Europe.
324 *Atmos. Pollut. Res.*, submitted.

325 Remer, L.A., Kaufman, Y.J., Tanré, D., Mattoo, S., Chu, D.A., Martins, J.V., Li, R.R., Ichoku, C., Levy,
326 R.C., Kleidman, R.H., Eck, T.F., Vermote E. and Holben, B.N. (2005). The MODIS aerosol
327 algorithm, products, and validation. *J. Atmos. Sci.* 62: 947–973.

328 Rienecker, M.M., Suarez, M.J., Todling, R., Bacmeister, J., Takacs, L., Liu, H.C., Sienkiewicz, M.,
329 Koster, R.D., Gelaro, R., Stajner, I. and Nielsen, J.E. (2008). The GEOS-5 data assimilation
330 system—documentation of versions 5.0.1, 5.1.0, and 5.2.0. Technical Report Series on Global
331 Modeling and Data Assimilation, Volume 27, NASA/TM-2008-104606.

332 Rienecker, M.M., Suarez, M.J., Gelaro, R., Todling, R., Bacmeister, J., Liu, E., Bosilovich, M.G.,
333 Schubert, S.D., Takacs, L., Kim, G.K., Bloom, S., Chen, J., Collins, D., Conaty, A., da Silva, A.,
334 Gu, W., Joiner, J., Koster, R.D., Lucchesi, R., Molod, A., Owens, T., Pawson, S., Pegion, P.,
335 Redder, C.R., Reichle, R., Robertson, F.R., Ruddick, A.G., Sienkiewicz, M. and Woollen, J. (2011).
336 MERRA: NASA’s Modern-Era Retrospective Analysis for Research and Application. *J. Clim.* 24:

337 3624–3648.

338 Schwartz, J. and Neas, L.M. (2000). Fine particles are more strongly associated than coarse particles with
 339 acute respiratory health effects in schoolchildren. *Epidemiology* 11: 6–10.

340 Smith, S.J., van Aardenne, J., Klimont, K., Andres, R.J., Volke, A. and Delgado Arias, S. (2011).
 341 Anthropogenic sulfur dioxide emissions: 1850–2005. *Atmos. Chem. Phys.* 11: 1101–1116.

342 Tsai, Y.I. and Cheng, M.T. (1999). Visibility and aerosol chemical composition near the coastal area in
 343 central Taiwan. *Sci. Total Environ.* 231: 37–51.

344 Tsai, Y.I. and Cheng, M.T. (2004). Characterization of chemical species in atmospheric aerosols in a
 345 metropolitan basin. *Chemosphere* 54: 1171–1181.

346 Tsai, Y.I. and Kuo, S.C. (2005). PM_{2.5} aerosol water content and chemical composition in a metropolitan
 347 and a coastal area in southern Taiwan. *Atmos. Environ.* 39: 4827–4839.

348 Turpin, B.J. and Lim, H.J. (2001). Species contribution to PM_{2.5} mass concentration: revisiting common
 349 assumptions for estimating organic mass. *Aerosol Sci. Technol.* 35: 602–610.

350 Vestreng, V., Myhre, G., Fagerli, H., Reis, S. and Tarrasón, L. (2007). Twenty-five years of continuous
 351 sulfur dioxide emission reduction in Europe. *Atmos. Chem. Phys.* 7: 3663–3681.

352 Wang, S. and Hao, J. (2012). Air quality management in China: issues, challenges, and options. *J.*
 353 *Environ. Sci.* 21: 2–13.

354 Willmott, C.J. (1982). Some comments on the evaluation of model performance. *Bull. Am. Meteorol.*
 355 *Soc.* 63: 1309–1313.

356 Xing, J., Pleim, J., Mathur, R., Pouliot, G., Hogrefe, C., Gan, C.M. and Wei, C. (2013). Historical gaseous
 357 and primary aerosol emissions in the United States from 1990–2010. *Atmos. Chem. Phys.* 13: 7531–
 358 7549.

359 Zhang, Q., He, K. and Huo, H. (2012). Cleaning China’s air. *Nature* 484: 161–162.

360 Zhao, B., Wang, S., Dong, X., Wang, J., Duan, L., Fu, X., Hao, J. and Fu, J. (2013). Environmental
361 effects of the recent emission changes in China: implications for particulate matter pollution and
362 soil acidification. *Environ. Res. Lett.* 8: 024031.

363 Zhao, Y., Zhang, J. and Nielsen, C.P. (2013). The effects of recent control policies on trends in emissions
364 of anthropogenic atmospheric pollutants and CO₂ in China. *Atmos. Chem. Phys.* 13: 487–508.

365

366 **Table 1.** Performance statistics for the ensemble of locations in Israel and Taiwan. AOC stands for
 367 “average observed concentration”.

	Israel	Taiwan
n	1,016,778	1,024,992
AOC ($\mu\text{g m}^{-3}$)	22.5	29.8
F	1.06	0.70
$\overline{\text{B}}$ ($\mu\text{g m}^{-3}$)	1.4	−8.8
SD-B ($\mu\text{g m}^{-3}$)	23.8	22.7
$\overline{\text{B}}_{log}$	0.07	−0.27
SD- B_{log}	0.66	0.83
R	0.56	0.27
R_{log}	0.50	0.30
FAC2	0.76	0.59

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369

370 **Table 2.** Average concentration simulated by MERRAero for the ensemble of locations in Fig. 1 over
 371 the study period.

Species	Israel		Taiwan	
	Average concentration ($\mu\text{g m}^{-3}$)	Proportion of PM _{2.5} concentration (%)	Average concentration ($\mu\text{g m}^{-3}$)	Proportion of PM _{2.5} concentration (%)
PM _{2.5}	24.0	—	20.9	—
(NH ₄) ₂ SO ₄	4.5	18.7	9.3	44.3
POM	1.3	5.4	3.0	14.1
BC	0.4	1.7	0.7	3.4
DS _{2.5}	13.9	57.9	2.0	9.3
SS _{2.5}	3.9	16.3	6.1	28.9

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374 **Table and figure captions**

375

376 **Table 1.** Performance statistics for the ensemble of locations in Israel and Taiwan. AOC stands for
377 “average observed concentration”.

378 **Table 2.** Average concentration simulated by MERRAero for the ensemble of locations in Fig. 1 over
379 the study period.

380 **Fig. 1.** Location of monitoring stations.

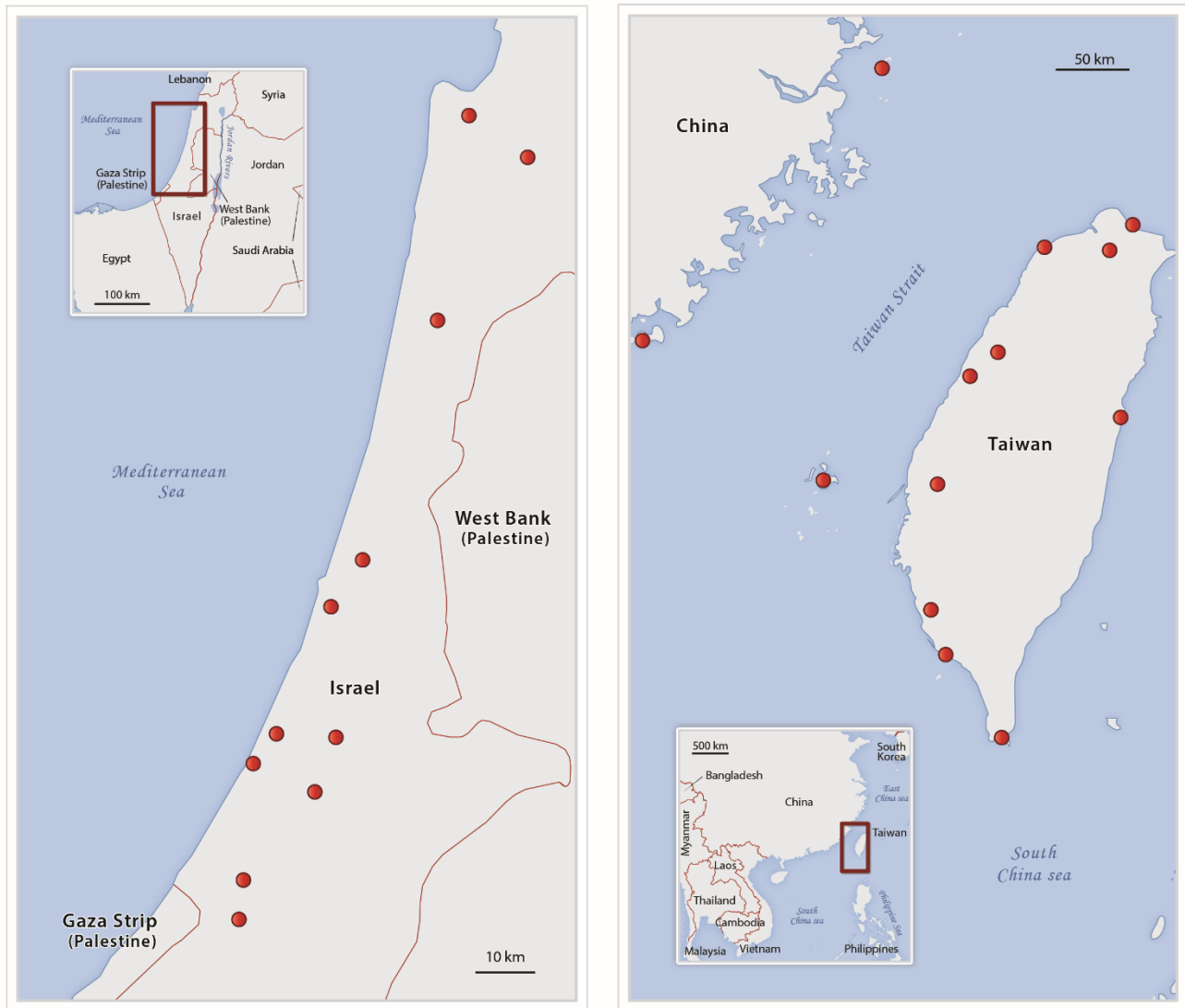
381 **Fig. 2.** Density scatter plot for (a) observed and simulated [PM_{2.5}], and (b) log transformed observed
382 and simulated [PM_{2.5}] for the ensemble of locations in Israel.

383 **Fig. 3.** (a) Yearly and (b) monthly average of [PM_{2.5}] observation, simulation and bias, and (c–d)
384 similarly for the SD and FAC2, for the ensemble of locations in Israel.

385 **Fig. 4.** Density scatter plot for (a) observed and simulated [PM_{2.5}], and (b) log transformed observed
386 and simulated [PM_{2.5}] for the ensemble of locations in Taiwan.

387 **Fig. 5.** (a) Yearly and (b) monthly average of [PM_{2.5}] observation, simulation and bias, and (c–d)
388 similarly for the SD and FAC2, for the ensemble of locations in Taiwan.

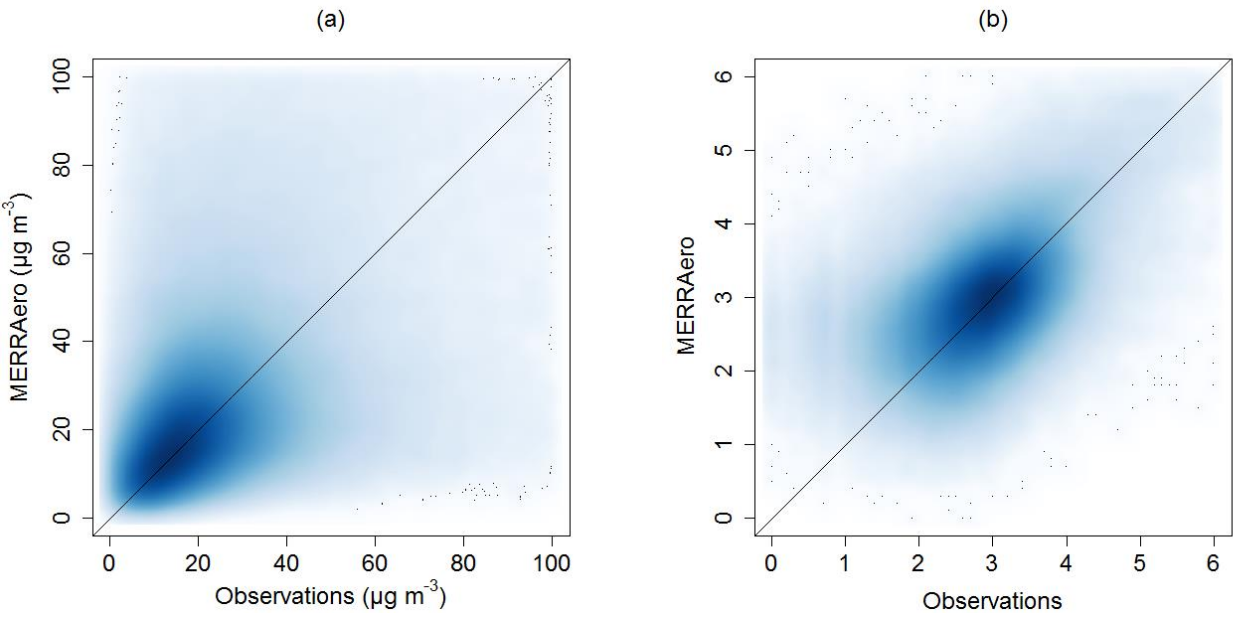
389



Realisation: Département de géographie, Université Laval, 2016

Fig. 1. Location of monitoring stations.

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395 **Fig. 2.** Density scatter plot for (a) observed and simulated $[PM_{2.5}]$, and (b) log transformed observed
396 and simulated $[PM_{2.5}]$ for the ensemble of locations in Israel.

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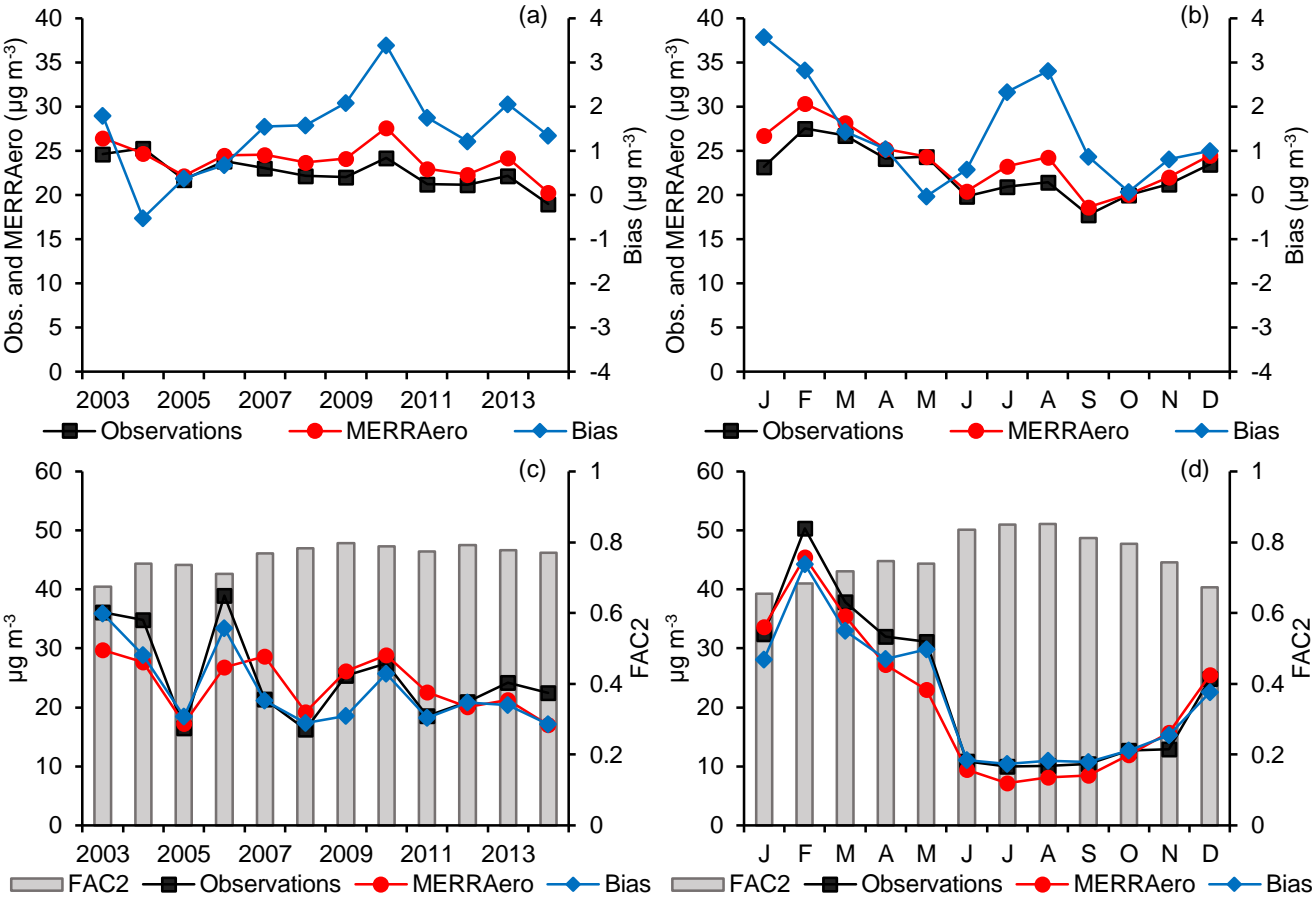
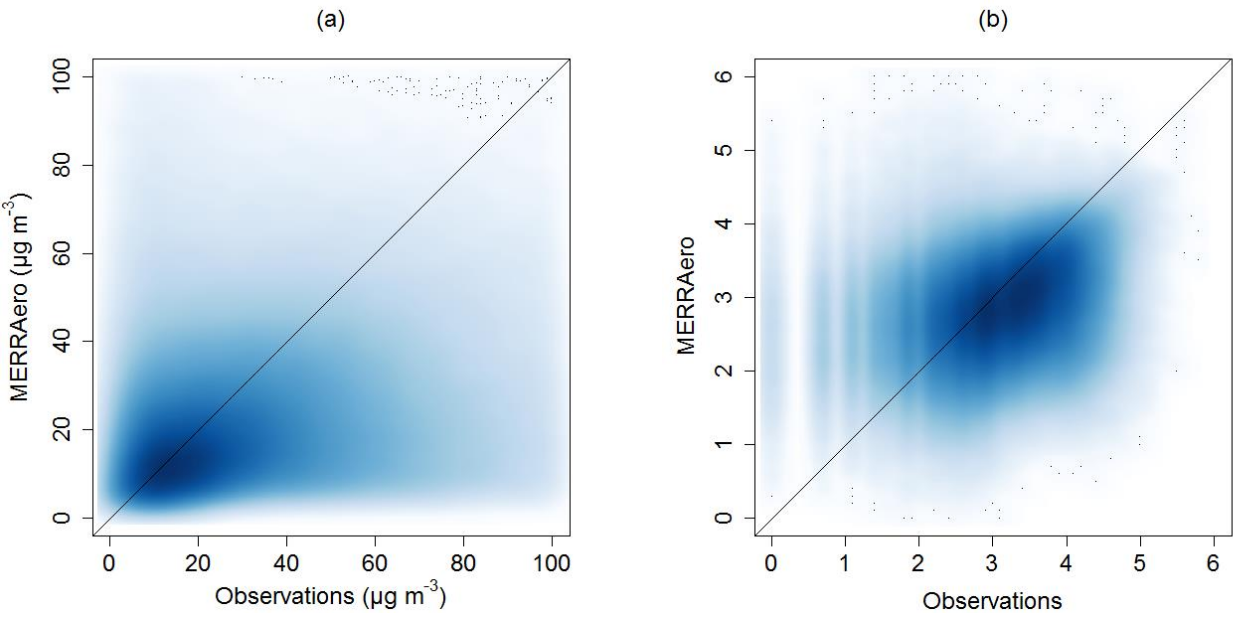


Fig. 3. (a) Yearly and (b) monthly average of [PM_{2.5}] observation, simulation and bias, and (c–d) similarly for the SD and FAC2, for the ensemble of locations in Israel.

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405

406 **Fig. 4.** Density scatter plot for (a) observed and simulated [PM_{2.5}], and (b) log transformed observed
407 and simulated [PM_{2.5}] for the ensemble of locations in Taiwan.

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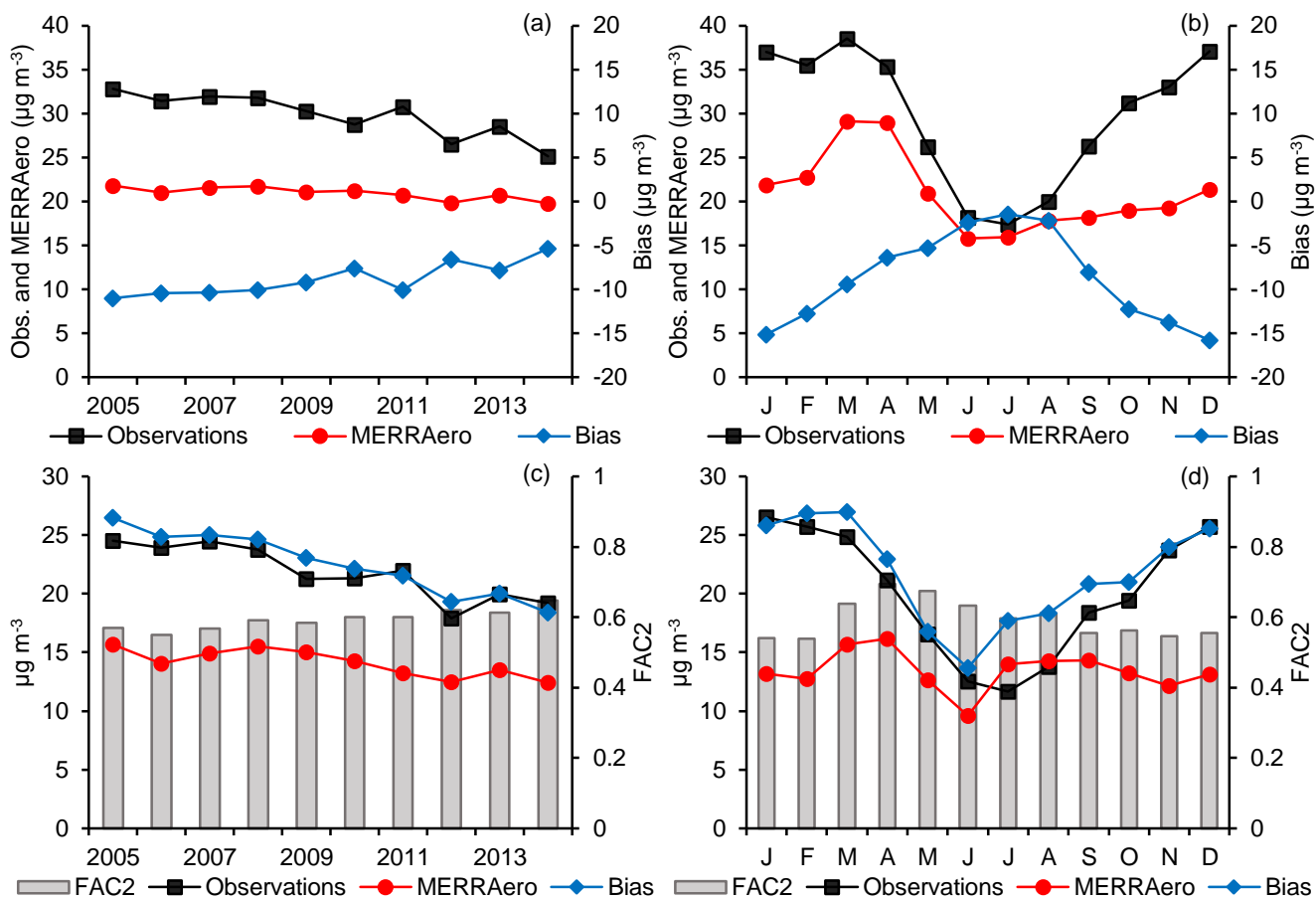


Fig. 5. (a) Yearly and (b) monthly average of [PM_{2.5}] observation, simulation and bias, and (c–d) similarly for the SD and FAC2, for the ensemble of locations in Taiwan.